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# High-Pressure Multi-Mbar Conductivity Experiments on Hydrogen: The Quest for Solid Metallic Hydrogen

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FY06 LDRD Final Report  
**High-Pressure Multi-Mbar Conductivity Experiments on Hydrogen:  
The Quest for Solid Metallic Hydrogen**

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## I Abstract

Ultra-dense hydrogen has long been the subject of intense experimental and theoretical research due to the fascinating physics which arises from this supposedly simple system. The properties of ultra-dense hydrogen also have important implications for planetary physics, since the interiors of the giant planets Jupiter and Saturn are believed to consist of cores of dense, metallic hydrogen. Finally, ultra-dense hydrogen is of direct programmatic interest, and multiple-shock compression experiments on hydrogen to the metallic state have stimulated the accelerated development of new hydrogen equation-of-state (EOS) models used for ICF and other applications.

The focus of our research has often been described as the "Holy Grail" of high-pressure physics research: The metallization of solid hydrogen. Metallic hydrogen has long been considered to be the prototypical system for the study of insulator-to-metal (I-M) transitions. Although metallic hydrogen ( $Z=1$ ) may superficially appear to be a very simple material, it is in fact an extremely challenging system for theoretical analysis due to the presence of large zero-point atomic motions and the complete absence of any core electrons. Thus, solid metallic hydrogen promises to be a fascinating material. Among its predicted properties is the possibility of being a high temperature superconductor with a critical temperature  $T_c$  of the order of  $\approx 100\text{K}$  [1]. The successful metallization of solid hydrogen would be a groundbreaking scientific discovery and open up new frontiers in science and possibly technology as well.

## II. Scientific Background

Ultra-high pressure hydrogen has been the subject of much experimental research by means of both dynamic (i.e., shock-wave compression) and static (i.e., diamond anvil cell) techniques. The most noteworthy recent work in this area was the LLNL discovery of *fluid* metallic hydrogen at high temperatures and pressures [2], which stimulated much renewed experimental and theoretical interest in dense hydrogen.

The discovery of *fluid* metallic hydrogen in no way diminishes the importance of achieving metallization in the solid state. Solid metallic hydrogen, with all of its possible exotic physical properties including high- $T_c$  superconductivity, still remains undiscovered. Research in this area has been hampered by the difficulty of performing electrical conductivity experiments with diamond anvil cells (DAC's) above a few hundred kbar and, as a result, most DAC studies on hydrogen have focussed on optical properties (e.g., optical absorption and Raman spectroscopy) or x-ray diffraction studies. Raman data [3] suggest that metallization may occur at around 300 GPa [4]. Optical absorption experiments indicate that the band gap of hydrogen is very small at high pressures, with the hydrogen sample appearing red at 305 GPa and then black at 320 GPa [5] (i.e., a band gap  $E_g < 2\text{eV}$  compared to an optical gap  $E_g = 15\text{ eV}$  for liquid hydrogen at 1 atmosphere).

The only Mbar electrical conductivity experiments on solid hydrogen are by one group which reported no measurable conductivity under pressures up to 210 GPa [6]. Based on all this information, electrical conductivity experiments at pressures of at least 300 GPa (3 Mbars) will be required for observing any conduction related to presence of solid hydrogen.

### III. Research Activities

The method most likely to succeed in observing electrical conductivity in solid hydrogen will be through the use of “designer” diamond anvils. Designer diamond anvils are specially fabricated diamond anvils with diamond encapsulated metal microcircuits (Figure 1). These anvils, developed as part of a joint LLNL and University of Alabama collaboration, are now in routine use in our research.

Designer diamond anvils are fabricated by starting with a standard, 1/3 carat diamond anvil and then lithographically fabricating a set of thin-film tungsten electrodes on the surface of the anvil. The electrodes extend from the lower sides of the anvil up to the tip of the anvil where the high-pressure sample is located. After probe fabrication, a high-quality epitaxial diamond film 10-50 nm thick is grown over the probes using microwave plasma chemical vapor deposition (MP-CVD) by our collaborators at the University of Alabama (Prof. Y.K. Vohra, Dept. of Physics). Finally, the rough as-grown diamond film on the tip of the anvil is precisely polished to a smooth, symmetrical shape so that it is suitable for high-pressure DAC experiments.

Part of our successful outcome of this LDRD project was modifying these anvils so that they are optimized for use up to multi-megabar pressures. We accomplished this goal by dramatically reducing the probe widths from 10  $\mu\text{m}$  down to 4  $\mu\text{m}$ . In addition, in order to minimize the diamond flat, we use only one electrical probe, which allows for the maximum compression of the gasket without electrically shorting out. In order to measure the electrical resistivity, we use the metallic gasket as the second lead. All of these small dimensions are necessary to reach the required goal of 3 Mbar.

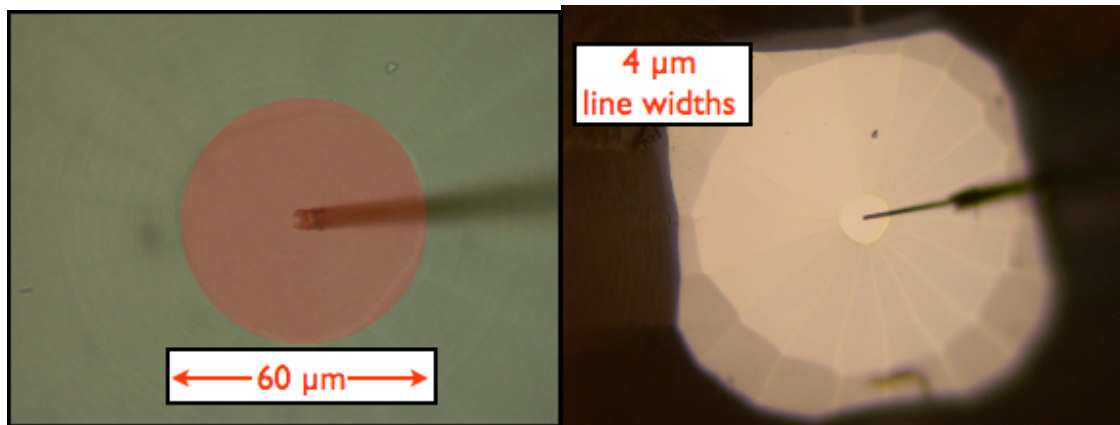


Figure 1. Designer diamond anvil with diamond encapsulated tungsten probes for high-pressure electrical conductivity experiments. The diamond has been optimized for multi-Mbar pressures by 1) reducing the flat to 60  $\mu\text{m}$  wide 2) using only one electrical lead, and 3) reducing the four electrical probe widths to 4  $\mu\text{m}$ .

The designer anvils were successfully integrated into our specially designed membrane diamond anvil cells (DACs) that were developed here at LLNL. These DACs use a gas membrane for applying load to the diamonds and thus pressurizing the sample captured between the diamond tips. We have used these membrane DACs extensively and find that they have many advantages over conventional screw- or lever-loaded DACs. The membranes uniformly apply load to the diamond, in contrast to screw-loaded DACs, and provide for very fine and precise control of the applied load, easily achieving pressures of several megabars for the newly fabricated designer diamonds. Our past experience using these cells has established a linear relationship between membrane pressure and sample pressure. Thus we can accurately increase to a desired pressure, and also monitor the deviation from linearity to diagnose when the diamonds are at imminent risk of failure. This capability will be useful in conducting experiments under precise pressure conditions and also avoiding unnecessary failure of the designer anvils.

During this project, we developed a method for loading hydrogen under high pressure ( $\approx 2$  kbar) at room temperature [7]. Hydrogen is an extremely compressible material, decreasing in molar volume by a factor of  $\sim 2$  upon increasing pressure to only 20 kbar [8]. In order to have a sufficiently large sample at the high pressures we are targeting, we must load the sample at as high a density as possible. The system safely compresses hydrogen gas up to 25 kpsi, which results in a hydrogen density within a percent of that of the liquid phase. The DAC can then be removed to begin measurements of the electrical resistivity.

#### **IV. Outcome**

The goal of this project, measuring the electrical resistivity of solid hydrogen, would have great implications for the scientific community if successful. However, the resistivity of solid hydrogen remained too large to be measured during our pressurizations. While we were able to optimize the designer diamond anvil technology for multi-megabar pressures, we were not able to overcome the diamond embrittlement caused by the hydrogen sample. In all thirteen of the attempts to load hydrogen into the DAC, cracks appeared in the designer diamond which resulted in cleaving the diamond into two to four pieces. The cracks appeared immediately after loading, and the highest pressure obtained before losing the sample was 1 Mbar, obtained for a single loading.

#### **V. Future Directions**

The greatest barrier in our successful pursuit of observing solid metallic hydrogen has been in the cracks that appear immediately after loading. While there is some evidence that pre-screening the diamonds through x-ray tomography could help in reducing the odds of a cracked diamond [9], this method can at best reduce the odds to an unimpressive 60% chance of remaining intact during an experiment. There are also discussions within the high pressure community that the source for the diamonds can be important in maintaining the diamond integrity. This may be due to the quality of the polishing process. Another avenue to pursue is to load the sample cryogenically, and maintain the DAC at cryogenic temperatures during the experiment.

## V. Summary

The pursuit of discovering solid metallic hydrogen falls into the “high risk/high payoff” category. During this project, we perfected designer diamond anvils for use at multi-megabar pressures by reducing the probe line-widths down to 4  $\mu\text{m}$  and reducing the diamond flat to 60  $\mu\text{m}$ . Designer diamond anvils are now optimized for future ultra-high pressure experiments. We were also successful in perfecting a system for loading gases into the DAC sample space. This method is now routinely used not only to load a gas sample, but also to load a hydrostatic pressure medium. However, after more than ten attempts at loading hydrogen into a DAC with designer diamonds, cracks would appear from within the center of the diamond, which resulted in the diamond prematurely breaking into multiple pieces. The path toward future success may lie in cryogenically loading the hydrogen sample.

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